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# Simultaneous high-speed measurement of temperature and lifetime-corrected OH laser-induced fluorescence in unsteady flames

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A means of performing simultaneous, high-speed measurements of temperature and OH lifetime-corrected laser-induced fluorescence (LIF) for tracking unsteady flames has been developed and demonstrated. The system uses the frequency-doubled and frequency-tripled output beams of an 80 MHz mode-locked Ti:sapphire laser to achieve ultrashort laser pulses (order 2 ps) for Rayleigh-scattering thermometry at 460 nm and lifetime-corrected OH LIF at 306.5 nm, respectively. Simultaneous, high-speed measurements of temperature and OH number density enable studies of flame chemistry, heat release, and flame extinction in unsteady, strained flames where the local fluorescence-quenching environment is unknown. © 2007 Optical Society of America

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Knowledge of both minor-species number density and temperature is essential for elucidating combustion chemistry mechanisms and validating numerical models of combustion. Tracking local flame extinction and global ignition–blowout is also important for the study of unsteady flames. Both temperature and the hydroxyl radical (OH) have been found to be sensitive markers of unsteady, strain-induced flame behavior, including local superequilibrium and extinction in nonpremixed flames [1]. In recent work, high-speed measurements of temperature and OH have been achieved using sum-frequency generation of ultraviolet (UV) radiation for diode-laser-based absorption spectroscopy [2]. While this approach achieves high data rates, data interpretation can be difficult in flames that are both inhomogeneous and unsteady because of the path-averaged nature of the absorption spectroscopy signal. High-repetition-rate planar laser-induced fluorescence (PLIF) of OH has also been achieved for four to eight sequential images [3], although the dependence of the OH fluorescence signal on local temperature and species concentration limits the quantitative nature of these measurements. Near flame extinction, for example, local fluorescence quenching can lead to underestimates of an approximate factor of 2 or 3 in measured OH concentration.

Picosecond time-resolved laser-induced fluorescence (PITLIF) is an alternative approach that offers high temporal and spatial resolution, as well as simultaneous determination of the local fluorescence quenching rate [4]. Unlike absorption spectroscopy, PITLIF data are acquired at single or multiple points, with spatial resolution defined by the focal diameter of the laser excitation beam and the confocal detection scheme. By tracking both OH fluorescence and fluorescence lifetime, PITLIF enables measurements that are independent of the local collisional environment [4,5]. Here we demonstrate simulta-

neous measurements of temperature and minor-species number density with a high spatial resolution and data rate by using Rayleigh scattering and PITLIF. This is achieved without substantially increasing the system complexity, and the timing of the Rayleigh scattering and PITLIF signals can be controlled with great precision. Correcting for the local collisional environment by using the recorded OH lifetime extends the applicability of this approach to spatially inhomogeneous combustion, and simultaneous detection of temperature provides an additional parameter of significant relevance for understanding combustion dynamics and unsteady chemical kinetics.

The laser source for PITLIF and Rayleigh scattering is a 2 ps, mode-locked Ti:sapphire laser with a repetition rate of 80 MHz, pumped by a continuous-wave frequency-doubled diode-pumped Nd:YVO<sub>4</sub> laser at 532 nm (Spectra-Physics Millennia) to achieve 1.6 W at 919.5 nm (see Fig. 1). This beam is frequency doubled in a  $\beta$ -barium borate (BBO) crystal to achieve 120 mW of laser power at 459.8 nm that is subsequently mixed with the fundamental at 919.5 nm in a lithium triborate (LBO) crystal for 20–30 mW of laser power at 306.5 nm. The beam is tuned to the bandhead of the OH A–X electronic transition in the (0,0) vibrational band, primarily exciting the  $R_1(6)$  to  $R_1(11)$  rotational lines. The averaged signal from this set of rotational lines was determined to be relatively temperature insensitive by using a LIFBASE simulation [6]. The beam is focused to a 140  $\mu$ m diameter within the flame by a 20 cm focal-length lens. The laser-induced fluorescence (LIF) signal is detected at 90° by using a pair of 15 cm focal length fused-silica lenses (5 cm diameter), a 0.1 m monochromator (Jobin-Yvon H10, 500  $\mu$ m exit slit, 1200 grooves/mm at 500 nm), and a photon-counting Hamamatsu HS5321 photomultiplier tube (PMT). The collection bandwidth of 10 nm ensures that there

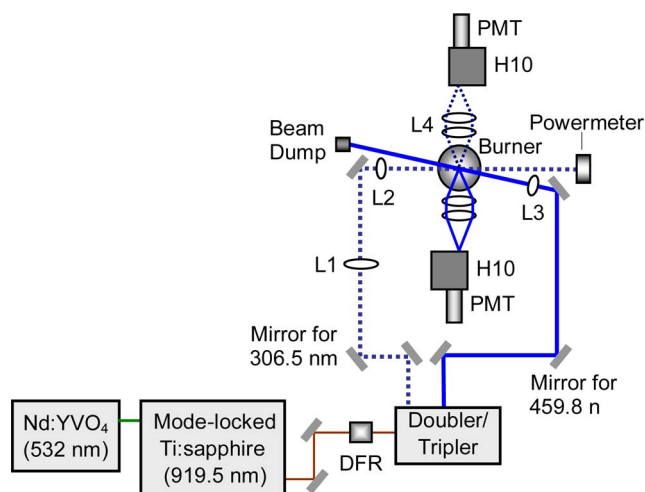


Fig. 1. (Color online) Optical layout for simultaneous Rayleigh-scattering and lifetime-corrected OH LIF. Symbols: PMT, photomultiplier tube; H10, 0.1 m monochromator; DFR, double Fresnel rhomb; L1, 1 m lens; L2, 20 cm lens; L3, 20 cm lens; L4, 15 cm lens.

are no line-dependent quenching effects on the OH signal. The 459.8 nm beam is focused to a 200  $\mu\text{m}$  diameter with a 20 cm focal-length lens and intersects the 306.5 nm beam at the probe volume. Rayleigh scattering at 459.8 nm is captured at 90° with a similar detection system consisting of a BK-7 lens pair, 0.1 m monochromator, and photon-counting PMT.

Each of the two PMT signals is amplified with a Philips Scientific 774 Quad Fast Risettime Amplifier, detected with two time-gated Philips Scientific 704 Quad 300 MHz discriminators, and digitized with a 100 kHz sampling rate, using four PC-based multichannel scalars (EG&G Ortec). Further details of the bin size and integration time controls are described in previous publications [4]. The temporal signal from each PMT is divided into three time-gated bins that are synchronized with the arrival of the laser by using a photodiode trigger from the Ti:sapphire laser. A timing diagram of the Rayleigh scattering and LIF signals from the two detection systems is shown in Fig. 2. The LIF system uses three channels to compute the fluorescence signal in a time sequence that minimizes Rayleigh scattering and allows determination of the fluorescence lifetime using a single-exponential model for the decay. A quenching-corrected OH-LIF measurement is obtained by extrapolating the fluorescence signal to a time just after the arrival of the laser excitation pulse when the effects of collisions are negligible. The Rayleigh-scattering signal is collected by the second detection system in a single 3.5 ns gated bin. Surface scattering is subtracted from the Rayleigh scattering signals in room air and the flame during postprocessing. Because the fuel mixture (22.1%  $\text{CH}_4$ , 33.2%  $\text{H}_2$ , and 44.7%  $\text{N}_2$ ) ensures the Rayleigh-scattering cross-sections of the reactants and products are nearly equal [7,8], the temperature is readily calculated from the ratio of the signal in room air to that in the flame and scaling by 295 K. The measurement standard deviations are 5% for the OH signal, 6% for OH

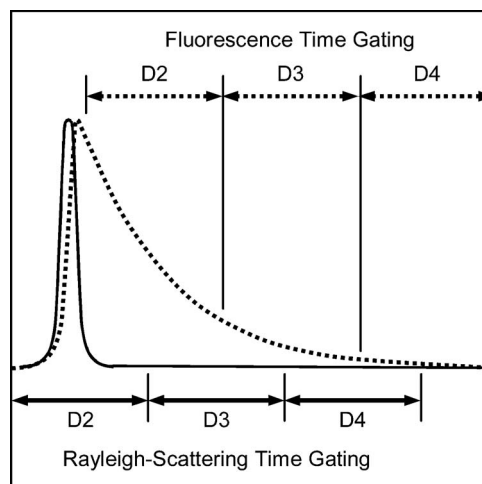


Fig. 2. Timing diagram for simultaneous detection of Rayleigh scattering and LIF. Channel D1 integrates over entire 12.5 ns period. Channels D2–D4 integrate over 3.5 ns bins to detect Rayleigh scattering and resolve fluorescence lifetime.

fluorescence lifetime, and 25% for temperature with a measurement bandwidth of 200–400 Hz (100 kHz sampling rate, 250–500 vortex/flame realizations). Strategies for significantly improving measurement bandwidth and signal-to-noise are discussed below.

Demonstration measurements were performed in an unsteady Rolon vortex/flame burner, pictured in Fig. 3 (left). As shown in Fig. 3 (right bottom), the fuel and oxidizer meet at a stagnation plane in the center of the burner. The steady stagnation flame is perturbed by a vortex injected from the bottom nozzle, such that the flame is deformed as shown 2.5 ms later in Fig. 3 (right top). The probe volume for the simultaneous temperature and lifetime-corrected LIF measurements is positioned to record the passage of the deformed flame.

The measurement of temperature and lifetime-corrected OH counts across the flame, as shown in Fig. 4, indicates the temperature layer is a factor of two wider than the OH layer, and the location of peak temperature is shifted by approximately one

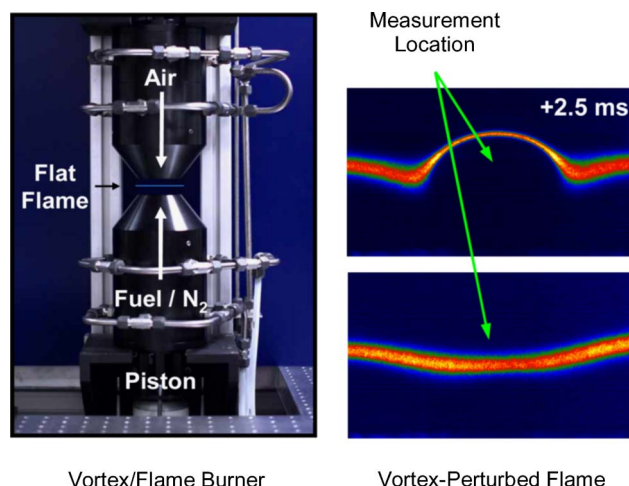


Fig. 3. (Color online) Rolon vortex/flame burner used to demonstrate simultaneous temperature and lifetime-corrected LIF measurements.

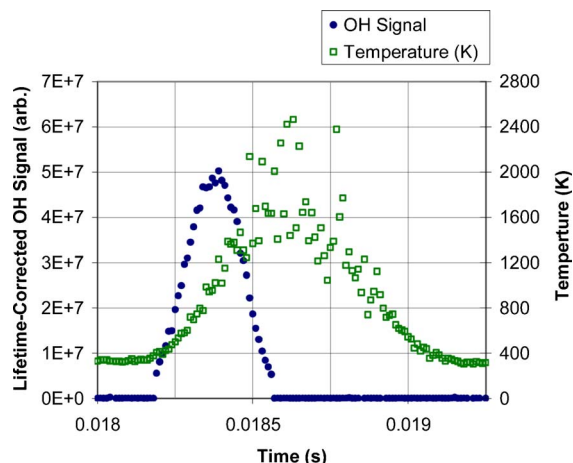


Fig. 4. (Color online) Simultaneous measurements of temperature and lifetime-corrected OH LIF during vortex/flame interaction. Passage of flame across probe volume reveals differences in width and peak of each profile.

OH-layer width from the location of peak OH. The absolute width of each layer can be deduced from knowledge of the flame velocity and vice versa. Such information regarding the relative distribution of temperature and flame intermediates, such as OH, is useful for validating numerical models of strain-induced chemistry in unsteady, propagating flamelets. The absolute accuracy of the temperature measurement, as deduced from the adiabatic flame temperature of the fuel mixture and the steady strain rate, is  $\sim 5\% - 7\%$ .

The OH-fluorescence lifetime recorded across the flame is shown with respect to the OH layer in Fig. 5 with a measured variation up to  $20\% - 25\%$ . As stated earlier, knowledge of the fluorescence lifetime allows one to correct for collisional quenching. Despite the large variation in temperature across the OH layer, the fluorescence lifetime is nearly symmetric and would artificially broaden the apparent OH profile as measured without lifetime correction. As the flame approaches extinction, the variation in quenching rate is expected to be more significant and

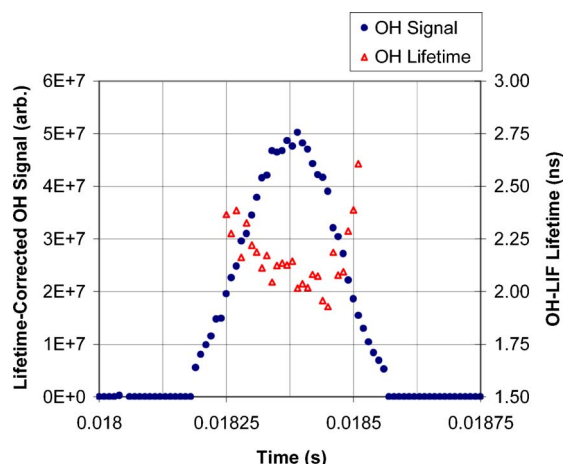


Fig. 5. (Color online) Simultaneous measurements of lifetime-corrected OH LIF with fluorescence lifetime variations of  $20\% - 25\%$  across the OH layer.

have a greater effect on the observed OH number density.

Based on the results of the proof-of-concept measurements here, it is possible to increase the signal-to-noise ratio by 2 orders of magnitude by (1) using the Ti:sapphire fundamental laser output for Rayleigh scattering, (2) increasing pump-laser energy and frequency-conversion efficiency, (3) utilizing larger collection optics, and (4) employing a more sensitive PMT. Recent tests of a more efficient frequency-conversion system, for example, produced over an order of magnitude increase in tripled laser energy for OH excitation. Taken together, such increases in signal-to-noise ratio yield a measurement bandwidth up to 2 orders of magnitude higher than the current work while reducing the measurement uncertainty up to fivefold. Rayleigh scattering, which yields weaker signals than OH fluorescence, would also be improved in high-pressure turbulent flames to be studied in future work.

While other systems exist for high-speed measurements of path-averaged temperature and minor-species number density, the current system enables single or multipoint measurements with high spatial resolution. This provides a means of performing simultaneous, high-speed measurements of temperature and lifetime-corrected LIF of minor species for tracking unsteady, spatially inhomogeneous flames. This unique capability enables the investigation of unsteady flames in which the collisional environment is unknown. The instrument setup is greatly simplified by using a single laser source to obtain temperature and minor-species number density, and the relative timing of the Rayleigh-scattering and LIF signals can be controlled with great precision. Finally, the wavelength tunability of the laser source allows electronic excitation of multiple species (e.g., OH, CH, or NO) with a single laser system.

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